Note on the hydrodynamic description of thin nematic films: strong anchoring model

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We discuss the long-wave hydrodynamic model for a thin film of nematic liquid crystal in the limit of strong anchoring at the free surface and at the substrate. Our aim is to clarify how the elastic energy enters the evolution equation for the film thickness; several models exist in the literature that result in qualitatively different behaviour. We consolidate the various approaches and show that the long-wave model derived through an asymptotic expansion of the full nemato-hydrodynamic equations with consistent boundary conditions agrees with the equation one obtains by employing a thermodynamically motivated gradient dynamics formulation based on an underlying free energy functional. As a result, we find that the elastic distortion energy is always stabilising in the case of strong anchoring. To support the discussion in the main part of the paper, an appendix gives the full derivation of the evolution equation for the film thickness via asymptotic expansion.

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I. INTRODUCTION

Thin films of nematic liquid crystals (NLC) have attracted attention over the years, as evidenced by a number of experimental and theoretical studies[1–10]. When thin nematic films are deposited on solid or liquid substrates, they often exhibit antagonistic anchoring at the free surface and at the substrate, i.e., the director orientation at the substrate is generally parallel to the substrate (planar anchoring) but at the free surface the director is orthogonal to the surface (homeotropic anchoring). As a consequence, the local director orientation changes across the film resulting in an elastic contribution to the energy that should not be neglected: such films are called hybrid films. Sometimes instabilities are observed that result in lateral periodic stripe patterns of the director orientation[1–6] and film height. However, this is only the case for thin films with thicknesses of several hundred nanometer and below; the wavelength of the stripe patterns diverges at an upper critical film thickness and so, for thicker films, only the usual defects of the nematic phases are observed[3,5]. Note that spinodal patterns have also been observed[8,11–13], normally, in the vicinity of nematic-isotropic or smectic-nematic phase transitions. In contrast, the stripe patterns are observed well inside the nematic region of the liquid crystal phase diagram.

In order to develop a theory for the behaviour of confined nematic liquid crystals, one may calculate the director orientation profile for a given static free surface. Typically, either a flat film or a periodically deformed state is considered. Such a given static geometry is then used to investigate the director field and to determine its stability. For an imposed flat film, an energy argument allows one to show that there exists a critical thickness[14]

$$h_c = \left| \frac{K}{A_+} - \frac{K}{A_-} \right|, \tag{1}$$

where K is the bulk elastic constant of the liquid crystal (in the one constant approximation) and A_+ and A_- are the anchoring strengths at the free surface and at the substrate, respectively. For thin films, with thickness $h \leq h_c$, the director profile is undistorted; the film is in the so-called planar (P) state and the director is aligned parallel to the anchoring angle at the interface with the stronger anchoring strength. For thick films, with film thickness $h > h_c$, the state that minimises the free energy is that where the director orientation changes continuously between the two anchoring directions as one moves across the film; this is the Hybrid-Aligned-Nematic (HAN) state introduced above and is the case for the strong anchoring situation considered here. If one assumes that the system is invariant in one direction across the surface on which the film is deposited, so that the film is effectively two-dimensional (2D), one finds that these states are

linearly stable. To confirm this assumption, much effort has gone into determining whether the film is laterally stable[1–3,10]. However, since the film geometry is imposed and static, such analyses can not account for a possible coupling of variations in film height and director orientation.

In alternative approaches, film thickness evolution equations for films of nematic liquid crystals are derived employing lubrication or long-wave hydrodynamic theory. This approach is used successfully to explore the dynamics of thin films of simple liquids under the influence of gravity or other body forces, and a variety of surface and interfacial forces[15–21]. Based on the long-wave approach, Ben Amar and Cummings[7] derived a model to describe the surface evolution of NLCs with strong anchoring in 2D settings that was later adapted to model 2D spreading droplets[22], spreading droplets with defects[23] and to account for three dimensional settings (3D)[24]. Another long-wave model was introduced by Carou et al. to study blade coating and cavity filling flows of NLC in 2D[25–27]. However, neither of these long-wave evolution equations agrees with models that use energy arguments, when it comes to ascertaining the effect of the elastic distortion energy on the film dynamics. Antagonistic anchoring is predicted to destabilise the film in Refs. 7, 22–24, but in Refs. 25–27 it is predicted to have no influence on the stability of the film. In Ref. 9, however, it is argued on physical grounds that the elastic energy is stabilising.

The purpose of this note is to clarify this issue by consolidating the hydrodynamic long-wave and energetic approaches in the case of layers of nematic liquid crystals with strong anchoring, and so to provide a solid basis for further investigations. The manuscript is organised as follows: The continuum theory of NLC, including the elastic energy and Ericksen-Leslie equations together with consistent boundary conditions, is given in Section II. Focusing on the 2D case, the long-wave approximation of the governing equations and boundary conditions is sketched in Section III while the full details are given in Appendix A. This allows the reader to easily reproduce our main findings. In Section IV, a thermodynamically motivated gradient dynamics formulation is employed to derive the evolution equation of a nematic film. The stability of the free surface is studied through a linear stability analysis. Finally, in Section V we compare the results of the two approaches and discuss the validity and limitation of the present model. The note concludes with an outlook on related problems that could be studied based on our results.

II. CONTINUUM DESCRIPTION OF NEMATIC LIQUID CRYSTAL

Nematic liquid crystals consist of rod-like molecules that have no positional order, but have long-range orientational order. Thus, the molecules are free to flow as a liquid, but still maintain their long-range directional order. The mean molecule alignment is described by the unit vector $\boldsymbol{n} = (n_1, n_2, n_3)^T$ where the superscript T denotes matrix transposition. Further notation conventions used here are presented in Appendix C.

Distortions of the director field result in a contribution to the free energy, that for NLC is known as the Frank-Oseen elastic energy and reads[28,29]

$$\begin{aligned} w_F = &\frac{1}{2}K_1(\nabla\cdot\boldsymbol{n})^2 + \frac{1}{2}K_2(\boldsymbol{n}\cdot\nabla\times\boldsymbol{n})^2 + \frac{1}{2}K_3(\boldsymbol{n}\times\nabla\times\boldsymbol{n})^2 \\ &+\frac{1}{2}(K_2+K_4)\nabla\cdot((\boldsymbol{n}\cdot\nabla)\boldsymbol{n} - (\nabla\cdot\boldsymbol{n})\boldsymbol{n}), \end{aligned} \tag{2}$$

where K_1, K_2 and K_3 are the splay, twist and bend elastic constants, respectively, and $(K_2 + K_4)$ is called the saddle-splay constant. Note that the saddle-splay term is often omitted since it does not contribute to the governing equations for problems invoking strong anchoring.

We use the one-constant approximation to simplify the problem. One assumes[28,29]

$$K \equiv K_1 = K_2 = K_3, \quad K_4 = 0. \tag{3}$$

and obtains the simplified energy density

$$w_F = \frac{K}{2}\nabla\boldsymbol{n} : (\nabla\boldsymbol{n})^T = \frac{K}{2}n_{l,k}n_{l,k} \tag{4}$$

that enters the nemato-hydrodynamic equations discussed next.

A. Ericksen-Leslie equation

The flow of NLC may be described by the Ericksen-Leslie equations[28–31]. The flow is incompressible, satisfying

$$\nabla\cdot\boldsymbol{v} = 0, \tag{5}$$

where $\boldsymbol{v} = (v_1, v_2, v_3)^T$ is the velocity field. The momentum balance equation is

$$\rho\frac{D}{Dt}\boldsymbol{v} = \nabla\cdot\boldsymbol{\sigma}, \tag{6}$$

where ρ is the density, $D/Dt = \partial/\partial t + \boldsymbol{v} \cdot \nabla$ is the material derivative, t is the time variable and $\boldsymbol{\sigma}$ is the stress tensor of the NLC. The stress tensor is defined as[28]

$$\boldsymbol{\sigma} = -p\,\boldsymbol{I} + \boldsymbol{\sigma}^E + \boldsymbol{\sigma}^V, \tag{7}$$

where p is the pressure, $\boldsymbol{I}$ is the identity tensor, $\boldsymbol{\sigma}^E$ is the elastic (Ericksen) stress tensor, defined by

$$\boldsymbol{\sigma}^E = -\frac{\partial w_F}{\partial \nabla \boldsymbol{n}} \cdot (\nabla \boldsymbol{n})^T, \tag{8}$$

and $\boldsymbol{\sigma}^V$ is the viscous stress tensor, written as

$$\left[\boldsymbol{\sigma}^V\right]_{ij} = \alpha_1 n_k n_p e_{kp} n_i n_j + \alpha_2 N_i n_j + \alpha_3 N_j n_i + \alpha_4 e_{ij} + \alpha_5 e_{ik} n_k n_j + \alpha_6 e_{jk} n_k n_i, \tag{9}$$

where

$$e_{ij} = \frac{1}{2}\left(v_{i,j} + v_{j,i}\right), \quad w_{ij} = \frac{1}{2}\left(v_{i,j} - v_{j,i}\right), \quad N_i = \frac{D}{Dt} n_i - w_{ik} n_k. \tag{10}$$

The α_i are constant viscosities.

The equation for the balance of angular momentum is written as (neglecting director inertia)

$$\nabla \cdot \left(\frac{\partial w_F}{\partial \nabla \boldsymbol{n}}\right) - \frac{\partial w_F}{\partial \boldsymbol{n}} + \boldsymbol{g} = \lambda \boldsymbol{n}, \tag{11}$$

where

$$[\boldsymbol{g}]_i = -\gamma_1 N_i - \gamma_2 e_{i,k} n_k, \quad \gamma_1 = \alpha_3 - \alpha_2, \quad \gamma_2 = \alpha_6 - \alpha_5. \tag{12}$$

Furthermore, λ is the Lagrange multiplier ensuring $|\boldsymbol{n}| = 1$.

Under the assumption of the one constant approximation, the Ericksen-Leslie equations, Eqs. (5), (6), and (11) simplify to

$$\nabla \cdot \boldsymbol{v} = 0, \tag{13a}$$

$$\rho \frac{D}{Dt} \boldsymbol{v} = -\nabla(p + w_F) - K\,\nabla \boldsymbol{n} \cdot \Delta \boldsymbol{n} + \nabla \cdot \boldsymbol{\sigma}^V, \tag{13b}$$

$$K \Delta \boldsymbol{n} + \boldsymbol{g} = \lambda \boldsymbol{n}, \tag{13c}$$

respectively, where we have used that

$$\boldsymbol{\sigma}^E = -K\,\nabla \boldsymbol{n} \cdot (\nabla \boldsymbol{n})^T, \tag{14}$$

and

$$\nabla \cdot (\nabla \boldsymbol{n} \cdot (\nabla \boldsymbol{n})^T) = \frac{1}{2} \nabla \left(\nabla \boldsymbol{n} : (\nabla \boldsymbol{n})^T\right) + \nabla \boldsymbol{n} \cdot \Delta \boldsymbol{n}. \tag{15}$$

As a result, the Ericksen-Leslie equations in the one constant approximation are given by Eq. (13) and need to be solved subject to appropriate boundary conditions.

a. Remark 1: Note that sometimes the stress tensor for NLC is written differently from Eq. (7), e.g., Ref. 32 uses

$$\tilde{\boldsymbol{\sigma}} = -(\tilde{p} + w_F)\,\boldsymbol{I} + \boldsymbol{\sigma}^E + \boldsymbol{\sigma}^V.$$

However, one may combine the two terms of the isotropic part of $\tilde{\boldsymbol{\sigma}}$ and define a modified pressure as $p = \tilde{p} + w_F$. Hence, with the exception of the modified pressure our derivations that follow are not affected.

b. Remark 2: Equation (13b) can be rewritten as

$$\rho\frac{D}{Dt}\boldsymbol{v} = -\nabla(p + w_F) + \nabla\boldsymbol{n}\cdot\boldsymbol{g} + \nabla\cdot\boldsymbol{\sigma}^V$$

by using Eq. (13c) together with $\nabla\boldsymbol{n}\cdot\boldsymbol{n} = \vec{0}$. This formulation is more popular in the literature since it only involves the first derivative of the director field.

1. Boundary conditions

We assume here that the NLC film sits on a solid substrate at $z = 0$ with the free surface (or film thickness) described by $z = h(x, y, t)$.

For the director field $\boldsymbol{n}$, we impose strong anchoring conditions such that the director is planar at the solid substrate and is homeotropic at the free surface. Specifically, we have

$$\boldsymbol{n}\cdot\boldsymbol{z} = 0, \quad \text{at } z = 0, \tag{16a}$$

$$\boldsymbol{n}\cdot\boldsymbol{t_i} = 0, \quad \text{at } z = h(x, y, t), \tag{16b}$$

where $\boldsymbol{z} = (0, 0, 1)^T$ and $\boldsymbol{t_i}$ are the surface tangent vectors,

$$\boldsymbol{t_1} = \frac{1}{\sqrt{1 + (\partial_x h)^2 + (\partial_y h)^2}}(1, 0, \partial_x h)^T, \quad \boldsymbol{t_2} = \frac{1}{\sqrt{1 + (\partial_x h)^2 + (\partial_y h)^2}}(0, 1, \partial_y h)^T. \tag{17}$$

For the velocity field $\boldsymbol{v}$, we assume no-slip at the solid substrate,

$$v_1 = v_2 = v_3 = 0, \quad \text{at } z = 0. \tag{18}$$

At the free surface, $z = h(x, y, t)$, we have the kinematic condition and balance of normal and tangential stresses. The kinematic condition is

$$v_3 = \partial_t h + v_1\partial_x h + v_2\partial_y h. \tag{19}$$

For normal stress, we assume that the jump across the interface is balanced by surface tension. That is,

$$\boldsymbol{k} \cdot [\boldsymbol{\sigma}]_i^n \cdot \boldsymbol{k} = 2\gamma H, \tag{20}$$

where $[\boldsymbol{\sigma}]_i^n = \boldsymbol{\sigma} - \boldsymbol{\sigma}^i$ and $\boldsymbol{\sigma}^i = -p_0 \boldsymbol{I}$ is the stress tensor of the air phase, p_0 is the atmospheric pressure, γ is the surface tension, H is the mean curvature and $\boldsymbol{k}$ is the surface normal vector

$$\boldsymbol{k} = \frac{1}{\sqrt{1 + (\partial_x h)^2 + (\partial_y h)^2}} \left(-\partial_x h, -\partial_y h, 1\right)^T. \tag{21}$$

For tangential stress, we assume that there is no jump at the interface

$$\boldsymbol{k} \cdot [\sigma]_i^n \cdot \boldsymbol{t_i} = 0. \tag{22}$$

That is, we assume that no tangential surface tension gradient exists, as is appropriate for strong anchoring. For the case where surface gradient exists, see Ref. 33.

III. LONG-WAVE HYDRODYNAMIC DESCRIPTION IN TWO DIMENSIONS

In this section we restrict attention to two space dimensions and focus on the long-wave approximation of the governing equations presented previously in Sec. II. The full details are given in Appendix A. The aim here is to study the contribution of nematic elasticity to the free surface evolution, and to distinguish results obtained using different scalings and boundary conditions.

Assume the flow is two dimensional and y-independent, so that the director field can be expressed as $\boldsymbol{n} = (\sin\theta, \cos\theta)^T$ where the angle θ is taken as the difference between the director orientation and the positive z-axis, as shown in Fig. 1(a), and the velocity field is $\boldsymbol{v} = (u, w)^T$. We introduce long-wave scalings to nondimensionalize the governing equations. The scalings are

$$(x, z) = (L\bar{x}, \delta L\bar{z}), \quad (u, w) = (U\bar{u}, \delta U\bar{w}), \quad t = \frac{L}{U}\,\bar{t}, \quad p = \frac{\mu U}{\delta^2 L}\,\bar{p}, \tag{23}$$

where U is the scale of fluid velocity, $\delta = H/L \ll 1$ is the ratio between the typical film thickness scale, H, and a typical lateral length scale, L. In addition, in order to focus only on the nematic elasticity, we approximate the nematic viscous stress tensor by its Newtonian equivalent, setting $\sigma_{ij}^V = 2\mu e_{ij}$, where $\mu = \alpha_4/2$.

A. Weak elasticity

Assuming that the elastic free energy is weak compared to the pressure, we can introduce the dimensionless number (Ericksen number)

$$\bar{K} = \frac{\delta \mu U L}{K}. \tag{24}$$

The leading order equations are then given by (after dropping the over-bars)

$$\partial_x p = \partial_z^2 u, \tag{25a}$$

$$\partial_z p = 0, \tag{25b}$$

$$K\, \partial_z^2 \theta = 0, \tag{25c}$$

$$\partial_x u + \partial_z w = 0. \tag{25d}$$

In addition, the leading order boundary conditions are

$$\theta(z=0) = \frac{\pi}{2}, \quad \theta(z=h) = 0, \tag{26}$$

$$p = p_0 - C\, \partial_x^2 h, \quad \partial_z u = 0, \quad \text{at } z = h, \tag{27}$$

where $C = \delta^3 \gamma / \mu U$ is the inverse capillary number.

It is easily seen that the velocity field and the director field are decoupled. The film thickness evolution equation is obtained as

$$\partial_t h + \partial_x \left(\frac{C}{3} h^3\, \partial_x^3 h \right) = 0, \tag{28}$$

and the director field satisfies

$$\theta = \frac{\pi}{2} \left(1 - \frac{z}{h} \right). \tag{29}$$

This corresponds to the approach taken in Refs. 25–27.

B. Moderate elasticity

Instead, if we introduce the Ericksen number as

$$\bar{K} = \frac{\mu U L}{K}, \tag{30}$$

the leading order equations are given by (after dropping the over-bars)

$$\partial_x \left(p + \frac{K}{2} \left(\partial_z \theta\right)^2 \right) = \partial_z^2 u, \tag{31a}$$

$$\partial_z p = 0, \tag{31b}$$

$$K\, \partial_z^2 \theta = 0, \tag{31c}$$

$$\partial_x u + \partial_z w = 0, \tag{31d}$$

with leading order boundary conditions

$$\theta(z=0) = \frac{\pi}{2}, \quad \theta(z=h) = 0, \tag{32}$$

$$p = p_0 - C\, \partial_x^2 h - K\left(\partial_z \theta\right)^2, \quad -K\left(\partial_x \theta\, \partial_z \theta + \left(\partial_z \theta\right)^2 \partial_x h\right) + \partial_z u = 0, \quad \text{at } z = h. \tag{33}$$

Under such a scaling, the director field is decoupled from the flow and is given by Eq. (29). The tangential stress boundary condition in Eq. (33) is then reduced to $\partial_z u(z=h) = 0$. We can therefore solve for the pressure and velocity field exactly. As a result, the film evolution equation is given by

$$\partial_t h + \partial_x \left(\frac{C}{3} h^3\, \partial_x^3 h - \frac{\widetilde{K}}{3}\, \partial_x h \right) = 0, \tag{34}$$

where $\widetilde{K} = \pi^2 K/4$.

In contrast, Ben Amar & Cummings[7] employ Eqs. (31, 32) and impose the normal stress balance assuming that the jump of the pressure is balanced by surface tension alone, as is appropriate for Newtonian fluids; that is, they use Eq. (27) instead of Eq. (33). As a result, they obtain an equation much like Eq. (34) but with the opposite sign for the elasticity term. This issue will be discussed later in Sec. V.

IV. GRADIENT DYNAMICS FORMULATION FOR A THIN FILM OF NEMATIC LIQUID CRYSTALS IN TWO DIMENSION

It was noted some time ago that the time evolution equation for the height of a thin Newtonian film on a solid substrate can be written in a variational form in situations where inertia can be neglected[21,34,35]. The evolution of the film thickness h follows a dissipative gradient dynamics governed by equation

$$\partial_t h = \partial_x \left[Q(h)\, \partial_x \left(\frac{\delta F}{\delta h} \right) \right], \tag{35}$$

where $\delta/\delta h$ denotes functional variation with respect to h. The resulting relaxation dynamics is governed by the free energy functional F with the mobility function $Q(h)$.

Such an approach may also be used to obtain the evolution equation for a NLC film in the limit of moderate elasticity discussed above in section III B. Restricting our attention again to a 2D geometry, we simplify the elastic distortion energy for the case of lateral long-wave distortions, i.e., we assume the scalings given in Eq. (23). The bulk elastic energy is to leading order

$$w_F = \frac{K}{2}\,(\partial_z\theta)^2. \tag{36}$$

We further assume that the director adjusts instantaneously to its steady state as compared to the fluid relaxation time, i.e., we assume $K = O(\mu U L)$. Then, the director field can be solved for exactly to obtain a linear profile as shown in Eq. (29) assuming strong planar anchoring at the solid substrate and strong homeotropic anchoring at the free surface. The corresponding director orientation across the film is sketched in Fig. 1(b).

As a result, the bulk elastic energy w_F (energy/volume) of the NLC can be rewritten as:

$$w_F = \frac{\widetilde{K}}{2h^2}. \tag{37}$$

The free energy functional is then expressed as

$$F = \int C\,ds + \int \left(\int_0^h w_F\,dz\right)\,dx \approx \int \left[C\left(1 + \frac{(\partial_x h)^2}{2}\right) + h w_F\right]\,dx, \tag{38}$$

where $ds \approx (1 + (\partial_x h)^2/2)\,dx$ is the approximated surface element. The evolution equation of the film is given in gradient dynamics formulation by introducing F into Eq. (35):

$$h_t = -\partial_x\left[Q(h)\,\partial_x\left(C\,\partial_x^2 h + \frac{\widetilde{K}}{2h^2}\right)\right], \tag{39}$$

where the mobility function $Q(h)$ can be obtained from the Poiseuille NLC flow, Eq. (A25). One should note that Eq. (39) and Eq. (34) are identical when $Q(h) = h^3/3$.

A. Linear stability analysis

To have a basic understanding of the elastic contribution to the stability of NLC free surface, we analyse the linear stability of a flat film, $h = h_0$. Assuming $h = h_0 + \xi$, $\xi \ll h_0$ in Eq. (39), to leading order we have

$$\partial_t\xi = Q(h_0)\left(-C\,\partial_x^4\xi + \frac{\widetilde{K}}{h_0^3}\,\partial_x^2\xi\right). \tag{40}$$

With the harmonic mode ansatz $\xi = \exp(ikx + \omega t)$ one obtains the dispersion relation

$$\omega = -Q(h_0)\left(C\,k^4 + \frac{\widetilde{K}}{h_0^3}\,k^2\right). \tag{41}$$

Note that the constants C, $\tilde{K}$ and the film height h_0 are always positive and therefore the growth rate ω is never positive for any wavenumber k. This implies that the elastic term is always stabilising and in the case of strong anchoring the flat film $h = h_0$ is always stable if only capillarity and elasticity are taken into account.

V. DISCUSSION AND CONCLUSION

We have presented several approaches to derive the evolution equation for free surface films of nematic liquid crystals with strong anchoring at both interfaces. We have demonstrated the consistency between the long-wave approximation model, Eq. (34) and Eq. (A24), and the model derived through a thermodynamically motivated gradient dynamics formulation, Eq. (39). Moreover, the elastic energy contribution acts in a stabilising manner in each of these models, consistent with the physically-motivated arguments of, e.g., Ref. 9 and 36.

In contrast, the long-wave models of Refs. 7, 22–24, which use an alternative normal stress balance, lead to qualitatively different results. The normal stress boundary condition in these papers neglects the contribution of the elastic stress tensor, which leads to a change in sign of the elastic contribution in the free surface evolution equation. A third approach used by Carou et al.[25–27] scales the nematic elasticity such that, to leading order, the free surface is unaffected by the elasticity, and one recovers the Newtonian thin film equation, Eq. (28).

One should note that the strong anchoring models presented here are only valid for rather thick films as noted in Sec. I. First, the main assumption of the model – the strong anchoring of the director at both interfaces – is only valid for $h \gg h_c$ where h_c is defined in Eq. (1). For moderate film thickness $h \approx h_c$ or even thinner, the surface anchoring energy has to be taken into account. This may be done via an ad-hoc amendment of the free surface anchoring condition in Eq. (32) (cf. the approach taken in Ref. 22, for the model with the 'Newtonian' normal stress balance). Alternatively it may be modelled via the variational approach that will be the subject of future work.

Second, as for isotropic liquids with film thickness below about 100 nm, long- and short-range effective intermolecular forces between the substrate and the free surface have to be taken into

account possibly through a Derjaguin or disjoining pressure that describes wettability effects[37]. For nematic liquid crystals the influence of van der Waals interactions has been discussed, e.g., in Ref. 8 and 38. Additional Casimir-type forces may be induced by fluctuations of the director orientation, most notably in very thin films with uniform director orientation, i.e., in the planar (P) state[38,39]. Note, however, that the notions “disjoining pressure” or “structural disjoining pressure” are used in Refs. 40–42 to denote the pressure contribution resulting from the elasticity of the liquid crystal, i.e., the last term in Eq. (39).

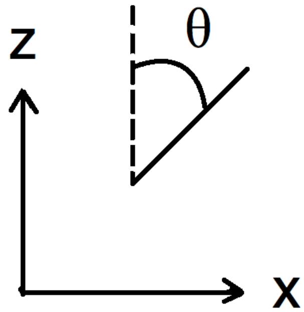
We would also like to point out that, within the present long-wave scalings (Eq. (23)), there is no distinction in the elastic energy whether the director is bent clockwise or counterclockwise. The two director profiles shown in Fig. 1(b) (on the right hand side and on the left hand side of the dashed (black) line) have exactly the same elastic energy, $\widetilde{K}/2h^2$. However, such a situation is still not allowed even though the elastic energy is continuous across the dashed (black) line. The director field is discontinuous and it breaks the long-wave assumption ($\partial_x^2\theta \ll \partial_z^2\theta$). A simple way to circumvent this was proposed in Ref. 23, whereby the discontinuity of Fig. 1(b) is smoothed out over a given range. More sophisticated models for real defects are needed. For instance, one may incorporate a description of the dynamics of the scalar order parameter related to the nematic-isotropic transition. Away from the phase transition it can be employed to model defects. Such a model would also allow one to tackle the structuring of films that occurs close to the nematic-isotropic transition[8,11–13].

In conclusion, we have clarified how the elastic contribution influences the free surface of a nematic film under the strong anchoring assumptions. Within the long-wave scalings, we have discussed two cases, corresponding to weak and moderate elasticity, respectively:

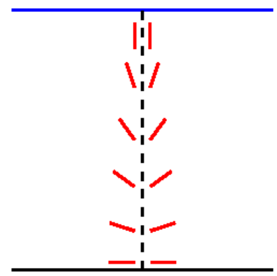
- $\frac{K}{\mu U L} = O(\delta)$: The bulk elasticity has only a minor influence on the free surface evolution. It does not affect the stability of a film. The evolution of the film and the director field are given by Eq. (A18) and Eq. (A20), respectively.

- $\frac{K}{\mu U L} = O(1)$: The strong antagonistic anchoring makes a significant contribution leading to a diffusion-like term in the film surface height evolution equation - see Eq. (39). Furthermore, the director always maintains its steady state, given by Eq. (29).

The models can be derived either from asymptotic expansion of the nemato-hydrodynamic equations or from a thermodynamically motivated gradient dynamics formulation. It is found that the elastic distortion energy is always stabilising.



(a)Coordinates



(b)Director orientation

FIG. 1. (Color online) The coordinates used in this manuscript are given in (a). The angle θ of the director is measured with respect to the positive z axis. In (b), we present two possible director profiles of a hybrid film. The molecules can bend either clockwise or counterclockwise across the film. The solid (blue) top curve indicates the free surface, the solid (black) bottom curve indicates the solid substrate, short (red) lines represent the orientation of the director field, and the dashed (black) line indicates the defect location.

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Appendix A: Long wavelength approximation of a thin film of NLC

1. Ericksen-Leslie equations in two spatial dimensions

Assume the flow is two dimensional and y-independent, then the director field can be expressed as $\boldsymbol{n} = (\sin\theta, \cos\theta)^T$ and the velocity field is $\boldsymbol{v} = (u, w)^T$. The elastic energy reduces to

$$w_F = \frac{K}{2}\left((\partial_x\theta)^2 + (\partial_z\theta)^2\right). \tag{A1}$$

Without fluid inertia, the linear momentum equations are then given by (from Eq. (13b))

$$\partial_x \left[p + \frac{K}{2} \left((\partial_x \theta)^2 + (\partial_z \theta)^2 \right) \right] = -K\, \partial_x \theta \, \left(\partial_x^2 \theta + \partial_z^2 \theta \right) + \partial_x \sigma_{11}^V + \partial_z \sigma_{12}^V, \tag{A2}$$

$$\partial_z \left[p + \frac{K}{2} \left((\partial_x \theta)^2 + (\partial_z \theta)^2 \right) \right] = -K\, \partial_z \theta \, \left(\partial_x^2 \theta + \partial_z^2 \theta \right) + \partial_x \sigma_{21}^V + \partial_z \sigma_{22}^V. \tag{A3}$$

(The viscous stress tensor, $\boldsymbol{\sigma}^V$, is defined in Appendix B, Eq. (B1).) For the angular momentum equation, Eq. (13c), one can eliminate the Lagrange multiplier λ by performing an inner product with the vector $\boldsymbol{n}^\perp = (\cos\theta, -\sin\theta)^T$. We then have

$$\begin{aligned} K(\partial_x^2 \theta + \partial_z^2 \theta) = \gamma_1 & \left[\dot{\theta} - \frac{1}{2}(\partial_z u - \partial_x w) \right] \\ & + \frac{\gamma_2}{2} \left[(\partial_x u - \partial_z w) \sin(2\theta) + (\partial_z u + \partial_x w) \cos(2\theta) \right]. \end{aligned} \tag{A4}$$

The continuity equation, Eq. (13a), is rewritten as

$$\partial_x u + \partial_z w = 0. \tag{A5}$$

a. Boundary conditions

In 2D, the boundary conditions for the director field, assuming strong anchoring at both interfaces (planar at the substrate and homeotropic at the free surface), are

$$\theta(z=0) = \frac{\pi}{2}, \quad \theta(z=h) = \cos^{-1}\left(\frac{1}{\sqrt{1 + (\partial_x h)^2}} \right). \tag{A6}$$

For the velocity field, we assume no-slip at the solid substrate,

$$u = w = 0, \quad \text{at} \quad z = 0. \tag{A7}$$

At the free surface we have the kinematic boundary condition

$$w = \partial_t h + u \partial_x h, \quad \text{at} \quad z = h, \tag{A8}$$

which can be combined with the incompressibility condition, Eq. (A5), to be

$$\partial_t h + \partial_x \left(\int_0^h u \, dz \right) = 0, \tag{A9}$$

or equivalently,

$$\partial_t h + \partial_x \left(\int_0^h \partial_z u \, (h - z) \, dz \right) = 0. \tag{A10}$$

(Note that the no-slip boundary condition, $u(z=0)=0$ was imposed in deriving Eq. (A10).)

For the balance of normal and tangential stresses, we first note that the stress tensor for a NLC film is written as

$$\boldsymbol{\sigma} = -p\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} - K\begin{bmatrix} (\partial_x\theta)^2 & \partial_x\theta\,\partial_z\theta \\ \partial_x\theta\,\partial_z\theta & (\partial_z\theta)^2 \end{bmatrix} + \begin{bmatrix} \sigma^V_{11} & \sigma^V_{12} \\ \sigma^V_{21} & \sigma^V_{22} \end{bmatrix} \tag{A11}$$

and the stress tensor of the air phase is $\boldsymbol{\sigma}^i = -p_0\boldsymbol{I}$. We assume that the jump in the normal stress is balanced by surface tension and the jump in tangential stress is zero. That is

$$\boldsymbol{k}\cdot[\boldsymbol{\sigma}]_i^n\cdot\boldsymbol{k} = \gamma\kappa, \quad \boldsymbol{k}\cdot[\boldsymbol{\sigma}]_i^n\cdot\boldsymbol{t} = 0, \quad [\boldsymbol{\sigma}]_i^n = \boldsymbol{\sigma} - \boldsymbol{\sigma}^i, \tag{A12}$$

where κ is the curvature, $\boldsymbol{k}$ is the normal vector at the free surface and $\boldsymbol{t}$ is the tangent vector at the free surface, defined as

$$\kappa = \frac{\partial_x^2 h}{(1+(\partial_x h)^2)^{3/2}}, \quad \boldsymbol{k} = \frac{1}{\sqrt{1+(\partial_x h)^2}}\left(-\partial_x h, 1\right)^T, \quad \boldsymbol{t} = \frac{1}{\sqrt{1+(\partial_x h)^2}}(1, \partial_x h)^T, \tag{A13}$$

respectively.

2. Non-dimensionalisation and long-wave approximation

We make the usual long-wave scalings to nondimensionalize the governing equations as shown in Eq. (23). Also we rescale the coefficients of nematic viscosity by the Newtonian equivalent, setting $\alpha_i = \mu\bar{\alpha}_i$ where $\mu = \alpha_4/2$. For the elastic constant, we assume $K = \epsilon\mu UL\bar{K}$ where ϵ is a parameter of order $o(1/\delta)$ that will be specified later.

The leading order equations are then given by (after dropping the over-bars)

$$\partial_x\left(p + \frac{\epsilon K}{2}(\partial_z\theta)^2\right) = -\epsilon K\,\partial_x\theta\,\partial_z^2\theta + \partial_z\left(q_1(\theta)\,\partial_z u\right), \tag{A14a}$$

$$\partial_z\left(p + \frac{\epsilon K}{2}(\partial_z\theta)^2\right) = -\epsilon K\,\partial_z\theta\,\partial_z^2\theta, \tag{A14b}$$

$$\epsilon K\,\partial_z^2\theta = -\delta q_2(\theta)\,\partial_z u, \tag{A14c}$$

$$\partial_x u + \partial_z w = 0, \tag{A14d}$$

where $q_1(\theta)$ and $q_2(\theta)$ are related to the viscous stress tensor, their full expressions are given later in Sec. B. The leading order boundary conditions are the kinematic boundary condition, Eq. (A10), with

$$\theta(z=0) = \frac{\pi}{2}, \quad \theta(z=h) = 0, \tag{A15}$$

$$p = p_0 - C\,\partial_x^2 h - \epsilon K\,(\partial_z\theta)^2, \quad -\epsilon K(\partial_x\theta\,\partial_z\theta + (\partial_z\theta)^2\,\partial_x h) + q_1(\theta)\,\partial_z u = 0, \quad \text{at } z = h, \tag{A16}$$

where $C = \delta^3\gamma/\mu U$ is the inverse capillary number.

a. Weak elasticity ($\epsilon = \delta$)

Assuming the elastic free energy is weak compared to the pressure, we can choose $\epsilon = \delta$. Observing that Eq. (A14b) reduces to $p_z = 0$ at leading order, one can solve the pressure exactly and the velocity is then determined by

$$\partial_z u(x,z) = \frac{C}{q_1(\theta)}\,(h-z)\,\partial_x^3 h. \tag{A17}$$

Hence, by using Eq. (A10), we obtain the film evolution equation as

$$h_t + C\,\partial_x\left(Q(h)\,\partial_x^3 h\right) = 0, \tag{A18}$$

where

$$Q(h) = \int_0^h \frac{(h-z)^2}{q_1(\theta)}\,dz. \tag{A19}$$

In addition, the director field satisfies

$$\partial_z^2\theta = \frac{C}{K}\,\frac{q_2(\theta)}{q_1(\theta)}\,(z-h)\,\partial_x^3 h \tag{A20}$$

with boundary conditions defined in Eq. (A15).

One can see that the nematic elasticity as well as viscosity only have influence on the mobility function Q, and thus have no influence on the stability of a free surface. This formulation has been studied extensively by Carou *et al.*[25–27] both analytically and numerically under the assumption of small director variation.

b. Moderate elasticity ($\epsilon = 1$)

On the other hand, if we have $\epsilon = 1$, Eq. (A14c) reduces to $\partial_z^2\theta = 0$ at leading order and hence the director field reaches a linear profile in z as shown in Eq. (29). Moreover, Eqs. (A14a) and (A14b) are simplified to

$$\partial_x\left(p + \frac{\widetilde{K}}{2h^2}\right) = \partial_z\left(q_1(\theta)\,\partial_z u\right), \tag{A21a}$$

$$\partial_z p = 0, \tag{A21b}$$

with boundary conditions at the free surface

$$p = p_0 - C\,\partial_x^2 h - \frac{\widetilde{K}}{h^2}, \quad \partial_z u = 0. \tag{A22}$$

We can therefore solve the pressure and velocity field as

$$p(x,z) = p_0 - C\,\partial_x^2 h - \frac{\widetilde{K}}{h^2}, \quad \partial_z u(x,z) = \left(-C\,\partial_x^3 h + \frac{\widetilde{K}}{h^3}\,\partial_x h\right)\left(\frac{z-h}{q_1(\theta)}\right). \tag{A23}$$

As a result, the film evolution equation is given by

$$h_t + \partial_x\left[Q(h)\left(C\,\partial_x^3 h - \frac{\widetilde{K}}{h^3}\,\partial_x h\right)\right] = 0, \tag{A24}$$

where $Q(h)$ can be evaluated explicitly as[7]

$$Q(h) = Q_0\,h^3, \quad Q_0 = \left(\frac{2}{\pi}\right)^3 \int_0^{\pi/2} \frac{\xi^2}{q_1(\xi)}\,d\xi. \tag{A25}$$

Appendix B: Viscous stress tensor of nematic liquid crystal in two dimension

The viscous stress tensor of NLC in 2D is written as

$$\begin{aligned}
\boldsymbol{\sigma}^V = {} & \alpha_1\left(\sin^2\theta\,\partial_x u + \cos^2\theta\,\partial_z w + \sin\theta\cos\theta(\partial_z u + \partial_x w)\right)\begin{bmatrix} \sin^2\theta & \sin\theta\cos\theta \\ \sin\theta\cos\theta & \cos^2\theta \end{bmatrix} \\
& + \alpha_2\left(\frac{D}{Dt}\theta - \frac{\partial_z u - \partial_x w}{2}\right)\begin{bmatrix} \sin\theta\cos\theta & \cos^2\theta \\ -\sin^2\theta & -\sin\theta\cos\theta \end{bmatrix} \\
& + \alpha_3\left(\frac{D}{Dt}\theta - \frac{\partial_z u - \partial_x w}{2}\right)\begin{bmatrix} \sin\theta\cos\theta & -\sin^2\theta \\ -\cos^2\theta & -\sin\theta\cos\theta \end{bmatrix} \\
& + \frac{\alpha_4}{2}\begin{bmatrix} 2\partial_x u & \partial_z u + \partial_x w \\ \partial_z u + \partial_x w & 2\partial_z w \end{bmatrix} \\
& + \frac{\alpha_5}{2}\begin{bmatrix} 2\sin^2\theta\,\partial_x u + \sin\theta\cos\theta(\partial_z u + \partial_x w) & 2\sin\theta\cos\theta\,\partial_x u + \cos^2\theta(\partial_z u + \partial_x w) \\ \sin^2\theta(\partial_z u + \partial_x w) + 2\sin\theta\cos\theta\,\partial_z w & \sin\theta\cos\theta(\partial_z u + \partial_x w) + 2\cos^2\theta\,\partial_z w \end{bmatrix} \\
& + \frac{\alpha_6}{2}\begin{bmatrix} 2\sin^2\theta\,\partial_x u + \sin\theta\cos\theta(\partial_z u + \partial_x w) & \sin^2\theta(\partial_z u + \partial_x w) + 2\sin\theta\cos\theta\,\partial_z w \\ \cos^2\theta(\partial_z u + \partial_x w) + 2\sin\theta\cos\theta\,\partial_x u & \sin\theta\cos\theta(\partial_z u + \partial_x w) + 2\cos^2\theta\,\partial_z w \end{bmatrix}.
\end{aligned} \tag{B1}$$

Similarly, the coupling term, $\boldsymbol{g}$, between the director and velocity field can be written as

$$\boldsymbol{g} = -\gamma_1\left(\dot{\theta} - \frac{\partial_z u - \partial_x w}{2}\right)\begin{bmatrix} \cos\theta \\ -\sin\theta \end{bmatrix} - \frac{\gamma_2}{2}\begin{bmatrix} 2\sin\theta\,\partial_x u + (\partial_z u + \partial_x w)\cos\theta \\ (\partial_z u + \partial_x w)\sin\theta + 2\cos\theta\,\partial_z w \end{bmatrix}. \tag{B2}$$

We also note that, within the long-wave scalings [Eq. (23)], to leading order we have

$$\sigma_{12}^V = \mu q_1(\theta)\,\partial_z u + O\left(\frac{\mu U}{L}\right), \quad \boldsymbol{n}^\perp \cdot \boldsymbol{g} = -\mu q_2(\theta)\,\partial_z u + O\left(\frac{\mu U}{L}\right), \tag{B3}$$

where $\mu = \alpha_4/2$, $\boldsymbol{n}^\perp = (\cos\theta, -\sin\theta)^T$ and

$$\mu q_1(\theta) = \frac{1}{2}\left[\alpha_4 + 2\alpha_1 \sin^2\theta\cos^2\theta + (\alpha_5 - \alpha_2)\cos^2\theta + (\alpha_3 + \alpha_6)\sin^2\theta\right], \tag{B4}$$

$$\mu q_2(\theta) = \frac{1}{2}\left[\gamma_1 - \gamma_2 \cos(2\theta)\right]. \tag{B5}$$

As an example, for Newtonian fluids, $q_1(\theta) = 1$ and $q_2(\theta) = 0$.

Appendix C: Notation conventions

For clarity, we list all the notations used. We write for a vector $\boldsymbol{n} = n_i$ or $\boldsymbol{m} = m_i$, for a tensor $\boldsymbol{\sigma} = \sigma_{ij}$ or $\boldsymbol{\kappa} = \kappa_{ij}$; as the superscript T denotes transposition one has $\boldsymbol{\sigma}^T = \sigma_{ji}$. Further, ϵ_{ijk} is the alternator. The notations for operators and products are $\nabla\boldsymbol{n} = n_{j,i}$, $\nabla\cdot\boldsymbol{n} = n_{k,k}$, $\nabla\times\boldsymbol{n} = \epsilon_{ilk}n_{k,l}$, $\nabla\cdot\boldsymbol{\sigma} = \sigma_{ik,k}$, $\boldsymbol{\sigma}\cdot\boldsymbol{\kappa} = \sigma_{ik}\kappa_{kj}$, $\boldsymbol{\sigma} : \boldsymbol{\kappa} = \sigma_{kl}\kappa_{lk}$, $\boldsymbol{\sigma}\cdot\boldsymbol{n} = \sigma_{ik}n_k$, $\boldsymbol{n}\times\boldsymbol{m} = \epsilon_{ilk}n_k m_l$, where '$,i$' denotes the partial derivative with respect to the ith component.

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